

A unified treatment of current-induced instabilities on Si surfaces

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We introduce a simple two-region model where the diffusion constant in a small region around each step on a vicinal surface can differ from that found on the terraces. Steady state results for this model provide a physically suggestive mapping onto kinetic coefficients in the conventional sharp-step model, with a negative coefficient arising from faster diffusion in the step region. A linear stability analysis of the resulting sharp-step model provides a unified and simple interpretation of many experimental results for current-induced step bunching and wandering instabilities on both Si(111) and Si(001) surfaces.

Si surfaces heated with a direct electric current undergo striking morphological changes induced by step bunching and step wandering instabilities. [1, 2] These phenomena are great interest not only because of possible applications for directed growth and nanofabrication, but also as physical examples of pattern formation on large (μm) length scales in a system driven far from equilibrium by a weak and externally controllable field.

We present here a simple model that describes the interplay between the externally driven diffusion of adatoms by the electric field, and the intrinsic modulations and anisotropy of the diffusion pathways and the attachment kinetics on terraces and steps. It provides a new interpretation of the kinetic coefficients used in traditional step models and shows that negative kinetic coefficients can arise from faster diffusion in the region around a step. Many features of the instabilities seen on Si surfaces can be understood from this perspective.

Step bunching is seen on vicinal Si(111) surfaces when the current is properly directed normal to the steps.[1, 2] The uniform step train is initially stable when the current flows in the opposite direction. This instability has a mysterious dependence on temperature, with three temperature ranges between about 850°C and 1300°C where the stable and unstable directions are reversed. Moreover, in temperature range II (about 1050°C to 1150°C) after heating for several hours with a current in the nominally stable (step-down) direction, the steps undergo a novel *wandering instability* with finite wavelength in-phase sinusoidal undulations in their positions.[2, 3]

Current-induced step bunching also occurs on Si(001) miscut along $\langle 110 \rangle$, but with some notable differences presumably arising from the alternating (1x2) and (2x1) dimer reconstructions on adjacent equilibrium terraces. At low temperatures step bunching is found for current normal to the steps in *both* directions, but involving paired double height steps. [2, 4, 5, 6]

The behavior in the lowest temperature range I of Si(111) is well described by a continuum diffusion model with nonequilibrium boundary conditions at sharp step edges [7] if one assumes that adatoms acquire a small

positive effective charge e^* and undergo biased diffusion [8] from a field-dependent force $\mathbf{F} = e^*\mathbf{E}$. However, it is not clear how to modify this picture to account for the reversals in the stable current direction at higher temperatures. Experiments [2] have ruled out the simplest explanation, a change of sign of the effective charge [9], and it seems likely that different boundary conditions are needed to describe the wandering instability. [10]

To gain some insight we introduce here a simple model that can give a more detailed description of processes occurring in the region around a step. The characteristic surface reconstructions seen on semiconductor surfaces strongly affect surface diffusion rates and pathways. We expect a different local reconstruction of bonds in the vicinity of a step. This suggests it could be profitable to view a step “dressed” by its local reconstruction as defining a *region* of finite width s (of a few atomic spacings a) where adatoms undergo effective diffusion with a diffusion constant D_s that can differ from D_t , the value found elsewhere on the terraces. [11] For Si(111) we can take D_s and D_t as isotropic and assume the step region has a fixed width s at a given temperature.

Thus a uniform vicinal Si(111) surface can be viewed as an array of two-region units, made up of the n th step region of width s and its neighboring lower terrace region, with width $l_t \gg s$. We assume that the straight steps extend along the y direction and the step index increases in the positive x (step down) direction, with x measured from the center of the step region. See Fig. (1).

The biased diffusion flux of adatoms with density c takes the form: $\mathbf{J}_\alpha = -D_\alpha \nabla c_\alpha + D_\alpha \mathbf{f} c_\alpha$, where $\alpha = (t, s)$ indicates the terrace or step regions and $\mathbf{f} \equiv \mathbf{F}/k_B T$. (We neglect evaporation and assume a constant positive effective charge.) We first consider the steady-state solutions that arise when the electric field is directed normal to the steps and let $f \equiv \mathbf{f} \cdot \hat{x}$. We can ignore the small effects of step motion on the steady state adatom density field and determine c by simply requiring $\nabla \cdot \mathbf{J}_\alpha = 0$ in each region, along with continuity of c and \mathbf{J} at the fixed boundary at $x = s/2$ between the step region and the lower terrace region. In almost all cases of physical interest, the field

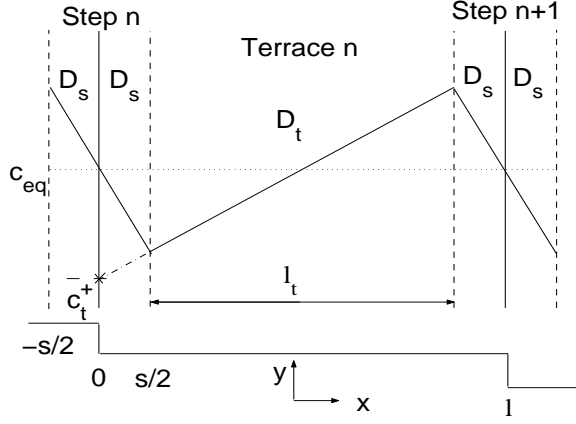


FIG. 1: Geometry and density profile of the two-region model. The sloping solid lines in the upper part of the figure give the steady state density profile as a function of distance x from the center of the left step region, denoted by the vertical dashed lines. Shown is a highly exaggerated profile for a downhill force and slower diffusion in the step region, yielding step bunching in range I in Si(111). Also illustrated with the dashed-dot line is the extrapolation of the terrace profile to the center of the step region, thus determining the parameter \bar{c}_t^+ in Eq. (3). The lower part of the figure gives a side view of sharp equilibrium steps and their associated step regions.

is sufficiently weak that fl_t and fs are much less than one, and the steady state profiles are piecewise linear.

In particular the steady state terrace density is

$$c_t^0(x) = c_{eq}^0 \left[1 - \frac{(l/2 - x)(R - 1)s f}{l + (R - 1)s} \right] \quad (1)$$

for $s/2 \leq x \leq l - s/2$, with a similar linear expression for $c_s^0(x)$. The (constant) adatom flux is given by

$$J_0(l) = \frac{D_t c_{eq}^0 f l}{l + (R - 1)s}. \quad (2)$$

Here $l \equiv l_t + s$ is the distance between the centers of adjacent step regions, $R \equiv D_t/D_s$ is a key dimensionless parameter that describes the relative diffusion rates of adatoms in the terrace and step regions, and c_{eq}^0 is the average equilibrium density when $f = 0$. This is also the density at the center of the step (and terrace) region, so one can view the step region as comprised of a classical local equilibrium sharp step at $x = 0$ surrounded by a symmetric local region of width s with a different diffusion constant, as illustrated in Fig. (1).

Equation (1) shows that the slope of the steady-state density profile is proportional to $(R - 1)f$. Thus, there are four general types of profiles, arising from a downhill force ($f > 0$) or uphill force ($f < 0$) combined with faster diffusion in the terrace region ($R > 1$) or in the step region ($R < 1$). We will use these results to make a precise connection between the physically suggestive parameters of the two-region model and appropriate boundary con-

ditions in an equivalent symmetric sharp-step model giving the same steady state terrace profiles. Both bunching and wandering instabilities can then be readily described using this standard framework.

The general continuum boundary condition in the sharp-step model assumes small deviations from local equilibrium and introduces linear *kinetic coefficients* k_{\pm} to relate \bar{c}_t^+ (or \bar{c}_t^-), the limiting lower (or upper) terrace adatom density at the step edge, to the associated terrace adatom flux into the step:

$$\mp \hat{n} \cdot \mathbf{J}_t^{\pm} = k_{\pm} (\bar{c}_t^{\pm} - c_{eq}). \quad (3)$$

Here $c_{eq} = c_{eq}^0 [1 + \Gamma\kappa]$ with Γ the capillary length (proportional to the step stiffness) and κ is the local step curvature. [7] For Si(111) experiments are consistent with a symmetric model where $k_+ = k_- = k$.

In the usual quasi-stationary approximation, the diffusion field with boundary conditions given by Eq. (3) is first calculated for fixed step positions, and then the step velocity is determined from the net local flux and mass conservation:

$$v_n \Delta c = \hat{n} \cdot [\mathbf{J}_t^- - \mathbf{J}_t^+] - \partial_{\tau} J_s. \quad (4)$$

Here $\Delta c = \Omega^{-1} \simeq a^{-2}$ is the change in areal density when an atom joins the solid, $J_s = -D_s \partial_{\tau} c_s + D_s (\hat{\tau} \cdot \mathbf{f}) c_s$ denotes a tangential or periphery diffusion flux along the step edge (the sharp-step analogue of parallel diffusion in the step region of the two-region model), and $c_s \simeq c_{eq}s$ gives the effective number of ledge atoms per unit step length with diffusion controlled by D_s rather than D_t .

It is natural to identify the terrace width in the appropriate sharp-step model with $l = l_t + s$ and to relate the limiting terrace density \bar{c}_t^+ in Eq. (3) to $c_t^0(0)$, the *extrapolation* of the two-region terrace profile in Eq. (1) to the center of the step region, as shown in Fig. (1). Relating parameters in discrete and continuum models by extrapolation is well known in other interface applications [12].

Using these results and the flux from Eq. (2) in Eq. (3), we find to lowest order in f our basic result:

$$\frac{D_t}{k} \equiv d = \frac{1}{2}(R - 1)s. \quad (5)$$

This equation relates the fundamental parameters R and s of the simplest two-region model to the kinetic coefficient k in an equivalent sharp-step model. [13]

Thus a positive kinetic coefficient k can arise from *slower diffusion in the step region* ($R > 1$), in accord with the usual picture of an attachment barrier in range I. Indeed the extrapolated profile in Fig. (1) corresponds exactly to the linear steady state profile analyzed in [7] if Eq. (5) is used to relate parameters in the two models. As $R \rightarrow 1$, we have $k \rightarrow \infty$, and we arrive at the local equilibrium boundary condition with $\bar{c}_t^{\pm} = c_{eq}$.

However, if diffusion is *faster* in the step region than in the terrace region ($R < 1$), we find a new regime with a

negative kinetic coefficient. Equivalently, the characteristic length $d = D_t/k$ is negative, but with $d \geq -s/2$.

The possibility of a negative kinetic coefficient in the presence of a Schwoebel barrier was first suggested by Politi and Villain, [14] but with no derivation or discussion of any physical consequences. We argue here that negative kinetic coefficients can play a key role in understanding current-induced instabilities on Si surfaces. This has quite different consequences than a model with permeable steps.[10] Consider a small perturbation $\delta x_n(y, t) \equiv x_n(y, t) - x_n^0 = \varepsilon e^{\omega t + i q y + i n \phi} + c.c.$ of the uniform step train. We report results for a linear stability analysis of the sharp step model in the physically relevant limits of weak fields ($fl \ll 1$) and long wavelengths ($ql \ll 1$). An instability arises from a positive $\omega = \omega_1(f, \phi) + \omega_2(q, f, \phi)$, where

$$\omega_1 = \Omega D_t c_{eq}^0 \frac{4df}{(l+2d)^2} (1 - \cos \phi), \quad (6)$$

and

$$\omega_2 = \Omega D_t c_{eq}^0 q^2 \left\{ -\Gamma \left[\frac{2(1 - \cos \phi)}{l+2d} + \left(l + \frac{s}{R} \right) q^2 \right] + f \left[\frac{2dl}{l+2d} + \frac{s}{R} \right] \right\}. \quad (7)$$

Step bunching is controlled by ω_1 . A pairing instability with maximum amplitude at $\phi = \pi$ is found for $df > 0$, or $(R-1)f > 0$, using the two-region model parameters. Thus, the profile illustrated in Fig. (1), produced by a step down current and slower diffusion in the step region, is unstable to step bunching, consistent with the usual interpretation [7] of range I in Si(111). But a similar unstable profile arises from a *step up current* ($f < 0$) along with a *negative* d or k .

ω_2 characterizes 2D step wandering. The first term in square brackets is always stabilizing and has its minimum value for in-phase wandering with $\phi = 0$. The next term, proportional to the field, has two contributions. The first, proportional to $D_t c_{eq}^0 df q^2$, describes a Mullins-Sekerka or Bales-Zangwill instability [15] induced by the terrace density field for $df > 0$. But as shown above, step bunching occurs under these same conditions. Wandering of the bunched steps is generally suppressed, as is seen experimentally in range I of Si(111).

However, the second contribution, proportional to $D_s c_{eq}^0 s f q^2$, represents an alternate and quite general mechanism for step wandering that is operative whenever there is a *downhill* force ($f > 0$). Downhill step perturbations are amplified by a field-driven downhill flux of adatoms along the step edge with steady state density per unit step length approximated by $c_s^0 = c_{eq}^0 s$. Using Eq. (5) we see that the last term in Eq. (7) is always positive even when $d < 0$ and the Mullins-Sekerka contribution is stabilizing.

Consider now the implications of these results for Si(111). The low temperature experiments are well explained by slower diffusion in the step region, consistent with the usual picture of an attachment barrier. [7] At higher temperatures in range II we suppose that changes in reconstruction could result in faster diffusion in the step region, implying a negative d or k in the sharp-step model. Bunching then is predicted for $f < 0$, and step wandering for $f > 0$, in agreement with experiments [2] and computer simulations [17] of such a model. The negative kinetic coefficient reverses the bunching direction, which allows the general wandering instability from a downhill force to be easily seen. Indeed $d < 0$ and $f > 0$ represents the *only* case where step wandering occurs with current in the opposite direction to that giving step bunching. Using parameters appropriate for Si(111) we find that the most unstable wavelength is of order μm , in qualitative agreement with experiment.

Since only relative diffusion rates are important, one could imagine the diffusion rates changing again at higher temperatures so that $d > 0$, possibly describing range III. In this scenario, the transitions between the different temperature ranges would be associated with local equilibrium behavior as R passes through unity, where no step bunching or wandering would be seen.

These ideas also provide an interpretation of electromigration results for the technologically important Si(001) surface miscut along $\langle 110 \rangle$. At equilibrium rather straight S_A steps that run parallel to the dimer rows of the upper A terrace alternate with much rougher S_B steps that run perpendicular to the dimer rows of the upper B terrace [16]. Moreover, diffusion parallel to the dimer rows is up to a thousand times faster at low temperatures [2]. For driven diffusion with a current normal to the steps we thus have $D_t^B \gg D_t^A$, and we expect that this difference will dominate the physics of current-induced instabilities of Si(001).

To apply our step-region ideas to this case, we imagine as before that a classical local equilibrium step resides in the center of each step region, but now let the downhill half-step region differ from the uphill half-step region, and assume that diffusion in each half-step region is similar to that in the nearest adjacent terrace. Defining $R^i \equiv D_t^i / D_s^i$, with $i = (A, B)$, we can let R^i differ from unity, thus generating asymmetric kinetic coefficients. We require only that $D_s^B \geq D_s^A$, which seems quite reasonable since $D_t^B \gg D_t^A$. Thus $D_s^B - D_s^A = \xi_s^{AB} (D_t^B - D_t^A)$ with $\xi_s^{AB} \geq 0$. Special cases of this model include classical local equilibrium steps where $R^A = R^B = 1$ and a symmetric step model where $D_s^B = D_s^A$.

Experiments show that when a direct current is applied to a configuration of alternating S_A and S_B steps, the steps move in opposite directions and step pairs form. With a downhill current one finds double height D_B steps (consisting of an upper S_B step and a lower S_A step with a very narrow A terrace trapped in between) separated

by wide B terraces; the equivalent configuration with D_A steps and wide A terraces is seen for an uphill current. On continued exposure to current, a step bunching instability of the double height steps is seen for current in *either* direction at low temperatures. [2, 6]

The initial step pairing can be most easily understood by calculating the velocity of steps in a configuration of equally spaced straight S_A and S_B steps. [18] Using the flux given by Eq. (2) with the appropriate values of D_t and Eq. (4) we find that the initial velocity of an S_B step can be written as $v_B = K_{AB}(D_t^B - D_t^A)f$, where K_{AB} is positive and symmetric in A and B . The initial velocity of an S_A step is given by the same formula when A and B are swapped and hence has the opposite sign. Thus, for positive f , B terraces grow and A terraces shrink. Given the great difference in D_t^B and D_t^A , this process will continue (as is shown by a general analysis with unequal terrace widths [19]) until the terraces have very different sizes, with the final width l' of the narrow A terrace in the D_B step probably controlled by step repulsions [18] (not taken into account in this version of our model).

To explain the continued bunching of the double-height steps, we use the two-region model, with major terraces separated by a double-height step, which we treat as a single effective step region. However a minor terrace now resides in the center of the effective step region, and we must take this into account in our extrapolation analysis leading to Eq. (5). We can proceed as before if we note that the effective equilibrium density in the center \hat{c}_{eq}^0 is linearly modified by the weak field from its value c_{eq}^0 at the “real” local equilibrium step near the lower boundary of the effective step region, so that $\hat{c}_{eq}^0 = c_{eq}^0[1 - f(l' + s)/2]$. Thus the analogue of Eq. (5) is [13]

$$\hat{d}^i \equiv \frac{D_t^i}{k^i} = \frac{1}{2} \left[R^i - \left(2 + \frac{l'}{s} \right) \right] s, \quad (8)$$

where \hat{d}^i denotes an effective double step parameter.

As shown above, with a step down current ($f > 0$) B terraces grow and D_B steps form. According to Eq. (6), step bunching occurs when $d\hat{d}^B > 0$. Thus continued bunching of the D_B steps requires that \hat{d}^B in Eq. (8) is greater than zero, or $R^B > 2 + l'/s$. (Note that a local equilibrium assumption [18] with $R^B = 1$ can give step pairing, but is inconsistent with further step bunching.) With step up current ($f < 0$) A terraces grow and D_A steps form. Continued bunching now requires that \hat{d}^A is *negative*, or $R^A < 2 + l'/s$. This inequality can be satisfied even if $R^A > 1$, and depending on the value of l' , could hold under rather general conditions. At higher temperatures we expect values of R^i closer to unity due to thermal fluctuations. If R^B becomes less than $2 + l'/s$ at some higher temperature, then Si(001) could exhibit behavior much like range II of Si(111), with bunching only for a step up current, due to the (effective) nega-

tive coefficient for the D_A step. Step wandering of the D_B steps from a step down current, suppressed by the bunching at lower temperatures, would also be expected.

Generalizations of these ideas and applications to experiments where the current is directed at an angle to the steps [6], along with comparison to results of Monte Carlo simulations will be presented elsewhere. [19] We are grateful to Ted Einstein, Oliver Pierre-Louis, and Ellen Williams for helpful discussions. This work was supported by the NSF MRSEC grant DMR 00-80008.

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